

PROGRESS REPORT

RESEARCH OBJECTIVE

The goals of the research effort are to develop new robust diagnostic tools for glass melt monitoring in a high-temperature, corrosive, radioactive environment, and to advance glass melt chemical modeling to make possible real-time melt characterization via the new sensor capability. Millimeter-wave technology will be applied to the simultaneous measurement of temperature, conductivity, and viscosity for the first time. This new sensor technology will make possible better process control to improve reliability and efficiency of waste glass melters. Also, it will provide new data for bridging the gap between theoretical glass melt models and their relationship to melter performance. The work is closely coupled to the needs of the Defense Waste Processing Facility (DWPF), West Valley Demonstration Project (WVDP), and vitrification efforts at Hanford, Oak Ridge, and Idaho sites. This research is a collaboration between the Massachusetts Institute of Technology (MIT) Plasma Science and Fusion Center (PSFC), the Pacific Northwest National Laboratory (PNNL), and the Savannah River Technology Center (SRTC).

RESEARCH PROGRESS AND IMPLICATIONS

As of the second year good progress has been made in parallel on the various research objectives. Laboratory experiments at MIT have established the feasibility for real-time monitoring of all three parameters, temperature, conductivity, and viscosity. Also a new capability for molten glass density measurements at high-temperature was discovered. A key milestone in the second year was a meeting¹ with Tank Focus Area representatives at MIT on December 7, 1999 to discuss monitoring priorities and the transfer of this technology to the TFA. Viscosity measurement was identified as the most important need. Experiments following this meeting demonstrated a strong correlation between millimeter-wave molten glass displacement measurements and viscosity. These results imply that robust new sensors for characterizing glass properties *in situ* are possible for improving control of present and future generation melters. Progress is briefly summarized below by the collaborating institution that led the particular developments described.

PSFC (MIT): Experiments continued in the second year with millimeter-wave instrumentation at a frequency of 137 GHz on a laboratory furnace. In collaboration with PNNL a number of different waveguide types, materials, and sizes were tested for interfacing the remote electronics with the molten glass. The two main waveguide types were internally corrugated metal guides and internally smooth non-conducting guides, which can propagate the efficient HE₁₁ mode. Inconel 690 was used for testing waveguides of the first type. Performance was good for up to 1150 °C but repeated use up to 1180 °C showed degraded performance due to surface corrosion. Silicon carbide (SiC) and mullite (3Al₂O₃·2SiO₂) were used to test waveguides of the second type. Though tests of high resistivity SiC samples indicated this would make a good dielectric waveguide, the vendor for this material could not produce a high resistivity tube. Mullite on the other hand proved to be a very effective millimeter-wave waveguide material. It has been successfully used in repeated measurements to 1300 °C, both above and immersed into the molten glass with millimeter-wave transmission efficiencies of 93% in the present setup. The implication of this result is that robust access for millimeter-wave sensors into glass melters in an oxidizing environment is now well established.

¹ Participants: Bill Holtzcheiter, Frank Thomas III, *from SRTC*; Tom Thomas, *from INEEL*; Glenn Bastiaans, *from Ames*; S. K. Sundaram, *from PNNL*; Paul Woskov, John Machuzack, Kamal Hadidi, and Paul Thomas, *from MIT*

A number of experimental runs with two different Hanford glasses have been carried out to develop the measurement techniques using both the Inconel and mullite guides. The measurement of temperature and emissivity has been shown. The feasibility of viscosity measurements has also been established by sealing the waveguide with a window and connecting to a pressurized source of gas. The rate of flow of molten glass in the waveguide, in response to a pressure transient, has been shown to be dependent on glass viscosity. Furthermore, the magnitude of the liquid glass displacement for a given pressure is dependent on the liquid density. Data for a Hanford glass mix over a temperature range of 1000 - 1300 °C and corresponding viscosity range of 3,000 – 100 Poise has been acquired. Millimeter-wave molten glass turbulence is a significant feature at viscosity below 200 Poise. The viscosity measurement development work is currently ongoing to optimize measurement methods and techniques, and to obtain additional data on other glasses.

PNNL: Work at PNNL has focused on molten glass conductivity – chemistry relationships and supporting the experiments at MIT.

Electrical Conductivity: A high-accuracy coaxial-cylinders technique was adapted successfully to measure the conductivity of waste glass melts. Characterization of 3 Hanford glasses, 6 DWPF glasses, and 5 privatization glasses was completed. Conductivity of the 2 Hanford glasses and 1 DWPF glass was predicted using 1) first-principle process-product model, developed by Jantzen² and 2) empirical mixture models, developed by Hrma et al.³. Preliminary comparison of the predictions with the measured value showed promise for developing a good conductivity-chemistry relationship. Further development of this relationship is in progress.

Redox: The coaxial-cylinders technique was modified to include a stabilized zirconia reference electrode. This modified probe will be used to measure redox of the waste glass melts from the glass test matrix, using square-wave voltammetry. A modified version of Rapidox[®] probe will also be used. These results will be compared.

SRTC: Research at SRTC has focused on the development of an advanced “Process Model” based control system that would use information from *in situ* measurements of millimeter wave emissions from molten glass to determine the glass composition. The current process control strategy in the Defense Waste Processing Facility uses analytical measurements of the compositions of the feed components along with an assessment of the uncertainties in the analysis and in the process variables such as batch volumes to ensure that acceptable glass will be made downstream from the blend tank. This is a feed forward control strategy. Direct measurement of product composition offers the possibility of feed back process control where the measured product properties are used to optimize the control of the system. The millimeter wavelength radiation profile from the glass would be fed into a process model that accounts for uncertainties in the measurements and system response to minimize the amount of frit added to the glass. Unlike the current control strategy, the new control strategy uses information about the composition of the blend tank, the melter feed tank, and glass already in the melter to determine an optimum glass blend. The glass produced by this strategy is potentially a more nearly optimal composition than it is currently possible to obtain. Future work will focus on quantifying the benefits of a feedback control system and will also consider measurements in the presence of disturbances such as temperature fluctuations to the system.

² C. M. Jantzen, “First Principles Process-Product Models for Vitrification of Nuclear Waste: Relationship of Glass Composition to Glass Viscosity, Resistivity, Liquidus Temperature, and Durability,” pp. 37-51 in Nuclear Waste Management IV, Editor: G. G. Wicks, D. F. Bickford, L. Roy Bunnell, Ceramic Transactions, Volume 23, The American Ceramic Society, Westerville, Ohio, USA, 1991.

³ P. R. Hrma, G. F. Piepel, P. E. Redgate, D. E. Smith, M. J. Schweiger, J. D. Vienna, and D. -S. Kim, “Prediction of Processing Properties for Nuclear Waste Glasses,” Ceramic Trans. 61, 505-513 (1995).